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On Stabilizing and Accelerating SCF Using ITP in Solving Kohn–Sham Equation

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Abstract. It is found that imaginary time propagation method can effectively deliver a convergent result in solving Kohn–Sham equation, but a sufficient long simulation is needed to reach an accurate enough result, while the self-consistent field iteration method for Kohn–Sham equation can be more efficient when it works, but it sometimes suffers from divergence. In this work, we take advantage of the convergence of imaginary time propagation method by generating a quality initial guess to improve the behavior of self-consistent field iteration. A number of numerical experiments successfully show that i). for those self-consistent field iterations which are sensitive to the initial guess, the results obtained from imaginary time propagation method make the iterations converge, and ii). generally, the convergence of self-consistent field iteration can be accelerated by imaginary time propagation method. It is shown that all-electron models can be resolved well with the proposed method.

AMS subject classifications: 35Q55, 65N30

Key words: Self-consistent field iteration, imaginary time propagation, finite element method, all-electron calculation.

1 Introduction

With the development of the hardware, as well as the requirements appeared in modern physical and chemical experiments, quality numerical simulations for all-electron Kohn–Sham models in density functional theory have been attracting more and more attention, please refer to [20, 26, 28, 30, 34] and references therein.

There are two popular approaches for solving Kohn–Sham model, i.e., the imaginary time propagation (ITP) method [3, 6–8, 14, 27], and the self-consistent field (SCF) iteration method [4, 5, 20, 26, 32, 33]. The implementation of ITP is quite simple. By introducing an

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imaginary time by Wick rotation, the complex-valued time-dependent Kohn–Sham equation is transformed to a real-valued time-dependent equation whose solution approaches asymptotically the ground state of the given system. Based on our numerical experience [25] which will also be illustrated in the context, it is noted that a sufficiently long simulation is needed by ITP method to obtain the ground state with an accurate enough result. As another widely-used method, SCF iteration method introduces a sequence of iterations to resolve the nonlinearity of the Kohn-Sham equation. More specifically, in each iteration, a generalized eigenvalue problem is derived with a given electron density, then the new electron density is generated by solving this generalized eigenvalue problem. The ground state of the given system is obtained when the iteration converges. As a brief comparison, the implementation of ITP method is simple, and the solving of eigenvalue problem is avoided. Although it may bring nontrivial challenges on solving eigenvalue problems, the SCF method could be more efficient when it works. Unfortunately, there are lots of evidence [24, 36, 37] to show the possible failures on the convergence of SCF iteration. The situation could become worse when all-electron Kohn–Sham models are considered where the mesh grid around the singularities should be dense enough to resolve the singularities. If a uniform mesh is adopted in the simulations, a great amount of degrees of freedom will be required to obtain an accurate result, which makes the computation less of efficiency. In this work, we follow the theoretical framework by Huang et al. [17] to derive a general mesh density function for any atomic or molecular system, and apply the result in generating the radial mesh in the numerical examples.

In solving the Kohn–Sham equation with a random initial guess on a nonuniform mesh, the following two issues may bring troubles in the convergence of SCF iteration: i) the condition number of the discretized Hamiltonian is too large, and ii) the initial guess is quite far away from the solution. On the one hand, the linear system becomes difficult to solve when the minimum mesh size $h_{\min}$ is small. This is due to the fact that the condition number of discretized matrix will become larger when $h_{\min}$ goes smaller [15]. On the other hand, the random initial guess could lead to the divergence of the SCF iteration. Due to the nonlinearity of the Kohn–Sham system, the quality of the initial guess plays an important role in the convergence of the iterative method. A bad initial guess which is far away from the convergence region will lead to divergent results, which can be observed from the numerical examples in this paper.

To improve the convergence of SCF, one could provide a good initial guess to the SCF iteration. Based on our numerical experience, it is found that ITP can provide an acceptable result rapidly in just a few steps even starting from a random initial guess. This motivates us to take the advantage of ITP to obtain a quality initial condition for the SCF iteration.

In this work, starting from a random initial guess, firstly the ITP method is applied to propagate the random initial guess for a few steps, and then the ITP solution is served as the initial guess of the SCF iteration to solve the Kohn–Sham equation. Due to the flexibility in handling the complex boundary condition and complicated computational domain, the finite element method is adopted in spatial discretization in this paper. It is noted